Supplementary information for Rotational self-diffusion in suspensions of charged particles: Simulations and revised Beenakker-Mazur and Pairwise Additivity methods

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S1 Revised Beenakker-Mazur method

We present here the details of our revised Beenakker-Mazur (BM) method in its application to rotational self-diffusion. The essential derivation steps are provided, with the focus set on the physical picture underlying the expansion in renormalized concentration fluctuations. The latter are commonly addressed as $\delta\gamma$ fluctuations.

The calculation of the short-time rotational self-diffusion coefficient on basis of Eq. (5) in the main text is not straightforward. The difficulty lies both in the calculation of the equilibrium probability distribution function, and of the $3N \times 3N$ rotational mobility matrix $\boldsymbol{\mu}^{rr}$. The latter problem part is made explicit when the 3×3 tensor elements of the mobility matrix are represented in the form of a hydrodynamic scattering series,

$$\boldsymbol{\mu}_{ij}^{rr}(\mathbf{R}_1 \ldots \mathbf{R}_N) = P_r T_{ij}(\mathbf{R}_1 \ldots \mathbf{R}_N) P_r^{\dagger}, \tag{S1}$$

with T_{ij} given by

$$\begin{aligned} T_{ij}(\mathbf{R}_1 \ldots \mathbf{R}_N) &= \delta_{ij} M(\mathbf{R}_i) \\ &+ (1-\delta_{ij}) M(\mathbf{R}_i) G(\mathbf{R}_i, \mathbf{R}_j) M(\mathbf{R}_j) \\ &+ \sum_{\substack{k=1, \\ k\neq i, k\neq j}}^{N} M(\mathbf{R}_i) G(\mathbf{R}_i, \mathbf{R}_k) M(\mathbf{R}_k) G(\mathbf{R}_k, \mathbf{R}_j) M(\mathbf{R}_j) \\ &+ \ldots . \end{aligned} \tag{S2}$$

The usage of scattering series in the context of suspensions is well established[1,2]. Not to interrupt unnecessarily our line of reasoning, we therefore refer to the section S2 for the explicit expressions for the hydrodynamic matrices M and G, and the definition of the projection operator P_r with its Hermitian conjugate $P_r^{\dagger}$ in Eq. (S1). It is worth while to note here, however, that the matrix G is related to the Green function tensor,

$$\mathbf{G}_0(\mathbf{r}) = \frac{1}{8\pi\eta r}(\mathbf{1} + \hat{\mathbf{r}}\hat{\mathbf{r}}), \tag{S3}$$

of the Stokes equations for an unbounded infinite fluid where $\hat{\mathbf{r}} = \mathbf{r}/r$. This so-called Oseen tensor describes the flow generated by a point force at the origin. Moreover, the matrix $M(\mathbf{R}_i)$ describes the hydrodynamic response of a single sphere with its center at position $\mathbf{R}_i$. Each term in Eq. (S2) is called a scattering sequence. Two examples of a scattering sequence are

$$M(\mathbf{R}_1) G(\mathbf{R}_1, \mathbf{R}_2) M(\mathbf{R}_2) G(\mathbf{R}_2, \mathbf{R}_1) M(\mathbf{R}_1) G(\mathbf{R}_1, \mathbf{R}_2) M(\mathbf{R}_2) \tag{S4}$$

and

$$M(\mathbf{R}_1) G(\mathbf{R}_1, \mathbf{R}_3) M(\mathbf{R}_3) G(\mathbf{R}_3, \mathbf{R}_2) M(\mathbf{R}_2) . \tag{S5}$$

Each scattering sequence is a succession of matrices M which scatter the flow, and matrices G which freely propagate the flow between scattering events. This physical interpretation of a scattering sequence is useful in our further discussion, and for the description of the physical idea underlying BM method.

Fluctuation expansion

In one of their first papers on short-time transport properties of suspensions[3], Beenakker and Mazur used the so-called fluctuation expansion. The physical idea behind this expansion is related to the resummation of a certain class of scattering sequences. BM based their considerations on the scattering series given by the Eq. (2.2) in their Ref.[3]. In the present analysis, we formulate the fluctuation expansion on basis of the scattering series given by Eq. (S2). Both approaches are equivalent but the presentation of the BM theory is more straightforward using our scattering series representation.

The scattering sequence in Eq. (S5) starts at particle 2 at position $\mathbf{R}_2$ and ends at particle 1 at position $\mathbf{R}_1$, with particle 3 acting as the intermediate. The scattering sequence in Eq. (S2), with the first propagators G starting from the particle 2 and the second one ending at the particle 1, can have all the other $N-2$ particles as intermediates, summing thus up to the expression

$$\sum_{i=3}^{N} M(\mathbf{R}_1) G(\mathbf{R}_1, \mathbf{R}_i) M(\mathbf{R}_i) G(\mathbf{R}_i, \mathbf{R}_2) M(\mathbf{R}_2) . \tag{S6}$$

If we treat this expression on the mean-field level by neglecting correlations between the particle positions, it is approximated by the volume integral

$$n \int d^3R_3 \, M(\mathbf{R}_1) G(\mathbf{R}_1, \mathbf{R}_3) M(\mathbf{R}_3) G(\mathbf{R}_3, \mathbf{R}_2) M(\mathbf{R}_2) \tag{S7}$$

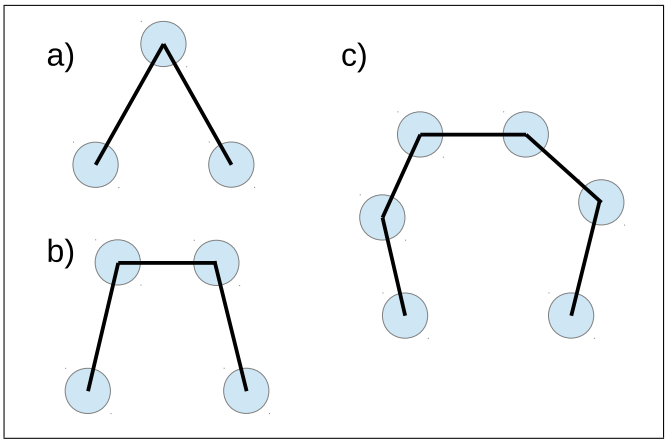

Figure S1 Schematic representation of scattering sequences resummed in the fluctuation expansion. a) Scattering sequence in Eq. (S7). b) Scattering sequence in Eq. (S8). c) Scattering sequence with five propagators.

with the number density n playing in a homogeneous system the role of the single-particle distribution function. The procedure of summing up similar scattering sequences involving three propagators G results in

$$n^2 \int d^3R_3 \int d^3R_4 M(\mathbf{R}_1) G(\mathbf{R}_1,\mathbf{R}_3) \times M(\mathbf{R}_3) G(\mathbf{R}_3,\mathbf{R}_4) M(\mathbf{R}_4) G(\mathbf{R}_4,\mathbf{R}_2) M(\mathbf{R}_2) . \quad \text{(S8)}$$

Notice that in the scattering sequences described by Eqs. (S7) and (S8), every reflection in the sequence is directed towards a new particle. This is schematically shown in Fig. S1. An example of a scattering sequence not satisfying this condition is given by Eq. (S4).

To proceed further, it is convenient to follow BM by introducing integral density kernels. Therefore, instead of $T_{ij}(\mathbf{R}_1 \ldots \mathbf{R}_N)$ in Eq. (S2), we consider its kernel density $\mathcal{T}$ defined by

$$\mathcal{T}(\mathbf{R},\mathbf{R}';\mathbf{R}_1 \ldots \mathbf{R}_N) = \sum_{i,j=1}^{N} \delta(\mathbf{R}-\mathbf{R}_i)\, T_{ij}(\mathbf{R}_1 \ldots \mathbf{R}_N)\, \delta(\mathbf{R}'-\mathbf{R}_j) , \quad \text{(S9)}$$

where $\delta(\mathbf{R})$ is the three-dimensional delta function. The kernel $\mathcal{T}$ is represented now as follows,

$$\mathcal{T} = \mathcal{M} + \mathcal{M}\tilde{G}\mathcal{M} + \mathcal{M}\tilde{G}\mathcal{M}\tilde{G}\mathcal{M} + \ldots \quad \text{(S10)}$$

where the densities $\mathcal{M}(\mathbf{R},\mathbf{R}';\mathbf{R}_1 \ldots \mathbf{R}_N)$ and $\tilde{G}(\mathbf{R},\mathbf{R}')$ are defined by

$$\mathcal{M}(\mathbf{R},\mathbf{R}';\mathbf{R}_1 \ldots \mathbf{R}_N) = \delta(\mathbf{R}-\mathbf{R}') \sum_{i=1}^{N} M(\mathbf{R}_i)\, \delta(\mathbf{R}-\mathbf{R}_i) , \quad \text{(S11)}$$

and

$$\tilde{G}(\mathbf{R},\mathbf{R}') = \begin{cases} 0 & \text{for } \mathbf{R}=\mathbf{R}' \\ G(\mathbf{R},\mathbf{R}') & \text{for } \mathbf{R}\neq\mathbf{R}' \end{cases} , \quad \text{(S12)}$$

respectively. In Eq. (S10), products of kernels such as $\mathcal{M}\tilde{G}$ appear which should be interpreted as:

$$\left[\mathcal{M}\tilde{G}\right](\mathbf{R},\mathbf{R}';\mathbf{R}_1 \ldots \mathbf{R}_N) \equiv \int d\mathbf{R}'' \mathcal{M}(\mathbf{R},\mathbf{R}'';\mathbf{R}_1 \ldots \mathbf{R}_N)\, \tilde{G}(\mathbf{R}'',\mathbf{R}') . \quad \text{(S13)}$$

Notice that Eq. (S10) is a short-hand notation since the kernel variables, the integral symbols and the position vectors of the particles are omitted. The full notation is used, in contrast, in Eq. (S13). It is worth noting that the only difference between $G(\mathbf{R},\mathbf{R}')$ and $\tilde{G}(\mathbf{R},\mathbf{R}')$ in Eq. (S12) is at $\mathbf{R}=\mathbf{R}'$. In introducing $\tilde{G}$ instead of G, BM have avoided the summation over the same particles since terms such as $M(\mathbf{R}_i)\tilde{G}(\mathbf{R}_i,\mathbf{R}_i)M(\mathbf{R}_i)$ are zero. The vanishing of $\tilde{G}(\mathbf{R}_i,\mathbf{R}_j)$ for $i=j$ excludes reflections to the same particle which are absent in the initial scattering series in Eq. (S2).

The sequence in Eq. (S7) is expressed in terms of the introduced kernels by

$$M(\mathbf{R}_1)\left[\tilde{G}\langle\mathcal{M}\rangle\tilde{G}\right](\mathbf{R}_1,\mathbf{R}_2)M(\mathbf{R}_2) , \quad \text{(S14)}$$

and the sequence in Eq. (S8) by

$$M(\mathbf{R}_1)\left[\tilde{G}\langle\mathcal{M}\rangle\tilde{G}\langle\mathcal{M}\rangle\tilde{G}\right](\mathbf{R}_1,\mathbf{R}_2)M(\mathbf{R}_2) . \quad \text{(S15)}$$

As throughout our paper, $\langle\ldots\rangle$ denotes the average with respect to the equilibrium configurational probability density function of N particles. Resummation of all scattering sequences for which each propagator connects to a different particle such as in the schematic scattering sequences in Fig. S1, yields

$$M(\mathbf{R}_1)\, G_{\langle\mathcal{M}\rangle}(\mathbf{R}_1,\mathbf{R}_2)\, M(\mathbf{R}_2) . \quad \text{(S16)}$$

Here $G_{\langle\mathcal{M}\rangle}$ is an "effective propagator", defined by

$$G_{\langle\mathcal{M}\rangle} = \tilde{G}\left(1-\langle\mathcal{M}\rangle\tilde{G}\right)^{-1} , \quad \text{(S17)}$$

which is the sum of a geometric series. The resummation of the above class of scattering sequences in the scattering series in Eq. (S10) leads to the so-called fluctuation expansion. The resummation can be performed by rewriting Eq. (S10) as follows

$$\mathcal{T} = \mathcal{M} + \mathcal{M}\tilde{G}\left[1-\mathcal{M}\tilde{G}\right]^{-1}\mathcal{M} . \quad \text{(S18)}$$

By adding and subtracting $\langle\mathcal{M}\rangle$, and using the operator identity

$$(1-A-B)^{-1} = (1-A)^{-1}\left[1-B(1-A)^{-1}\right] , \quad \text{(S19)}$$

for two operators A and B, we obtain the fluctuation expansion of the propagator $\tilde{G}\left[1-\mathcal{M}\tilde{G}\right]^{-1}$ as

$$\tilde{G}\left[1-\mathcal{M}\tilde{G}\right]^{-1} = \tilde{G}\left[1-(\mathcal{M}-\langle\mathcal{M}\rangle)\tilde{G}-\langle\mathcal{M}\rangle\tilde{G}\right]^{-1} \quad \text{(S20)}$$

$$= \tilde{G}_{\langle\mathcal{M}\rangle}\left[1-(\mathcal{M}-\langle\mathcal{M}\rangle)\tilde{G}_{\langle\mathcal{M}\rangle}\right]^{-1} . \quad \text{(S21)}$$

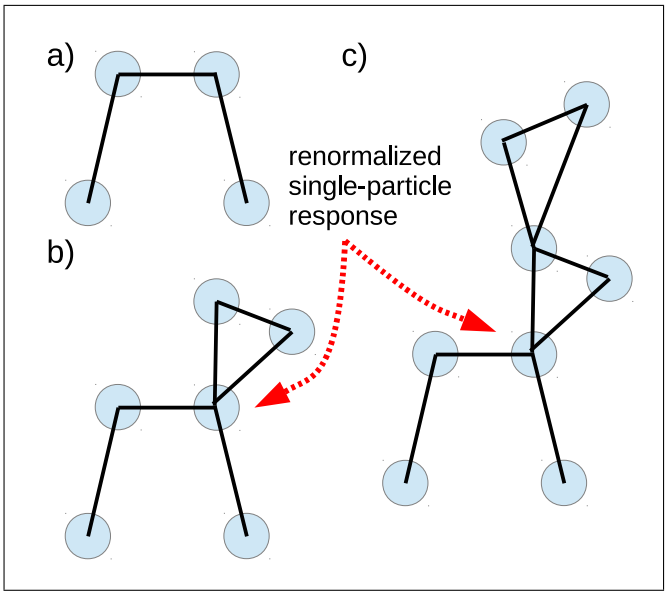

Figure S2 Schematic representation of scattering sequences resummed in a renormalized fluctuation expansion invoking the renormalized propagator in Eq. (S25). a) Scattering sequence without renormalization of the single-particle response (present also in the fluctuation expansion). b) and c) Scattering sequence with the single-particle operator M being renormalized.

When this expression is inserted in Eq. (S18), the fluctuation expansion result

$$\mathcal{T} = \mathcal{M} + \mathcal{M}\tilde{G}_{\langle\mathcal{M}\rangle}\left[1-(\mathcal{M}-\langle\mathcal{M}\rangle)\,\tilde{G}_{\langle\mathcal{M}\rangle}\right]^{-1}\mathcal{M}\,, \tag{S22}$$

for the kernel $\mathcal{T}$ is obtained.

Renormalized fluctuation expansion

The truncation of the fluctuation expansion for the translational mobility matrix has led to an approximate method of calculating the short-time translational self-diffusion coefficients[3]. However, the results by this second-order fluctuation expansion were found by BM to be unsatisfactory. Therefore, in subsequent work, BM performed another resummation which has led to the renormalized fluctuation expansion[4].

The scattering sequences illustrated in Fig. S1 for which each propagator links two different particles are resummed in the fluctuation expansion. In the renormalized fluctuation expansion, BM resummed similar scattering sequences using now a renormalized single-particle operator M_R. Namely, instead of the bare M operators, there appear now 'rings' built of scattering sequences with renormalized operators. This is illustrated schematically in Fig. S2. BM refer to these structures as 'ring self-correlations', which are scattering structures of the following form

$$\mathcal{M}_R = \mathcal{M}\left(1-G^s_{\langle\mathcal{M}_R\rangle}\mathcal{M}\right)^{-1}, \tag{S23}$$

where the integral operator $G^s_{\langle\mathcal{M}_R\rangle}$ is defined by

$$G^s_{\langle\mathcal{M}_R\rangle}(\mathbf{R},\mathbf{R}') = \begin{cases} G_{\langle\mathcal{M}_R\rangle}(\mathbf{R},\mathbf{R}') & \text{for } \mathbf{R}=\mathbf{R}' \\ 0 & \text{for } \mathbf{R}\neq\mathbf{R}' \end{cases}, \tag{S24}$$

while the renormalized propagator $G_{\langle\mathcal{M}_R\rangle}$ is of the form

$$G_{\langle\mathcal{M}_R\rangle} = \tilde{G}\left(1-\langle\mathcal{M}_R\rangle\,\tilde{G}\right)^{-1}. \tag{S25}$$

The derivation of the expansion in renormalized density fluctuations is omitted here for it involves only simple algebraic manipulations which are given in the works of BM[4–6]. The most important result of this derivation is the expansion of the operator $\mathcal{T}$ in renormalized fluctuations, $\mathcal{M}_R-\langle\mathcal{M}_R\rangle$, according to

$$\mathcal{T} = \mathcal{M} + \mathcal{M}G_{\langle\mathcal{M}_R\rangle}\left[1-(\mathcal{M}_R-\langle\mathcal{M}_R\rangle)\,\tilde{G}_{\langle\mathcal{M}_R\rangle}\right]^{-1}\mathcal{M}_R\,. \tag{S26}$$

The operator $\tilde{G}_{\langle\mathcal{M}_R\rangle}$ in the above expansion is defined by

$$\tilde{G}_{\langle\mathcal{M}_R\rangle}(\mathbf{R}_1,\mathbf{R}_2) = G_{\langle\mathcal{M}_R\rangle}(\mathbf{R}_1,\mathbf{R}_2) - G^s_{\langle\mathcal{M}_R\rangle}(\mathbf{R}_1,\mathbf{R}_2)\,. \tag{S27}$$

It is also worth quoting the intermediate result

$$\begin{aligned}&\tilde{G}\left(1-\mathcal{M}\tilde{G}\right)^{-1}\mathcal{M}\\ &= G_{\langle\mathcal{M}_R\rangle}\left[1-(\mathcal{M}_R-\langle\mathcal{M}_R\rangle)\,\tilde{G}_{\langle\mathcal{M}_R\rangle}\right]^{-1}(\mathcal{M}_R-\langle\mathcal{M}_R\rangle)\\ &+ G_{\langle\mathcal{M}_R\rangle}\left[1-(\mathcal{M}_R-\langle\mathcal{M}_R\rangle)\,\tilde{G}_{\langle\mathcal{M}_R\rangle}\right]^{-1}\langle\mathcal{M}_R\rangle\,,\end{aligned} \tag{S28}$$

used in the renormalized fluctuation expansion of the translational and rotational self-diffusion coefficients discussed in the following.

Renormalized fluctuation expansion for translational self-diffusion

Before considering the renormalized fluctuation expansion of the rotational self-diffusion coefficient, we consider first translational self-diffusion which was investigated earlier by Beenakker and Mazur. The short-time translational self-diffusion coefficient D^t is expressed by the following formula

$$D^t = \frac{k_BT}{3}\lim_{\infty}\left[\mathrm{Tr}\left\langle\frac{1}{N}\sum_{i=1}^{N}\boldsymbol{\mu}^{tt}_{ii}(\mathbf{R}_1\ldots\mathbf{R}_N)\right\rangle\right], \tag{S29}$$

in analogy with Eq. (5) in the main text for the rotational self-diffusion coefficient. The above formula has been re-expressed by BM in terms of kernels according to

$$D^t = \frac{k_BT}{3}\mathrm{Tr}\left(P_t dP_t^{\dagger}\right), \tag{S30}$$

where the matrix d is defined as follows:

$$d = nM(\mathbf{R}) + M(\mathbf{R}) \lim_{\infty} \left\langle \tilde{G} \left(1 - \mathcal{M}\tilde{G}\right)^{-1} \mathcal{M} \right\rangle (\mathbf{R},\mathbf{R}) \,. \quad \text{(S31)}$$

The operator P_t whose explicit form is given in section S2 projects on the translational components of the matrix d. This should be compared with the Eq. (3.16) in Ref[3]. The operator expression in Eq. (S28) inserted into the above expression results in the renormalized fluctuation expansion of d. Up to second order in the renormalized fluctuations, $\mathcal{M}_R - \langle \mathcal{M}_R \rangle$, this expansion reads

$$d = d^{(0)} + d^{(1)} + d^{(2)} + \ldots, \quad \text{(S32)}$$

where

$$\begin{aligned} d^{(0)} &= nM + M\left[G_{\langle \mathcal{M}_R \rangle} \langle \mathcal{M}_R \rangle\right](\mathbf{R},\mathbf{R}) \,, & \text{(S33)} \\ d^{(1)} &= 0 \,, & \text{(S34)} \\ d^{(2)} &= d_2^{(2)} + d_3^{(2)} \,, & \text{(S35)} \end{aligned}$$

and

$$\begin{aligned} d_2^{(2)} &= M\Big[G_{\langle \mathcal{M}_R \rangle} \Big\langle (\mathcal{M}_R - \langle \mathcal{M}_R \rangle) \tilde{G}_{\langle \mathcal{M}_R \rangle} \\ &\quad \times (\mathcal{M}_R - \langle \mathcal{M}_R \rangle) \Big\rangle\Big](\mathbf{R},\mathbf{R}) \,, & \text{(S36)} \\ d_3^{(2)} &= M\Big[G_{\langle \mathcal{M}_R \rangle} \Big\langle (\mathcal{M}_R - \langle \mathcal{M}_R \rangle) \tilde{G}_{\langle \mathcal{M}_R \rangle} \\ &\quad \times (\mathcal{M}_R - \langle \mathcal{M}_R \rangle) \Big\rangle \tilde{G}_{\langle \mathcal{M}_R \rangle} \langle \mathcal{M}_R \rangle \Big](\mathbf{R},\mathbf{R}) \,. & \text{(S37)} \end{aligned}$$

The truncation approximation,

$$d \approx d^{(0)} + d^{(1)} + d^{(2)} \,, \quad \text{(S38)}$$

constitutes along with Eq. (S30) and Eqs. (S32)-(S37) the second-order renormalized fluctuation expansion approximation for the short-time translational self-diffusion coefficient.

Renormalized fluctuation expansion for rotational self-diffusion

The extension of the renormalized fluctuation expansion method to short-time rotational self-diffusion was made first by Treloar and Masters[7]. This extension is straightforward in our formalism, since Eq. (5) for D^r is similar in structure as Eq. (S30) for D^t. The only difference appears in the invoked projectors, i.e. instead of the projector P_t on the translational components the projector P_r on the rotational components is used in Eq. (5). Since the renormalized fluctuation expansion has been introduced in the previous section on the general level of the d matrix, the extension from translational to rotational self-diffusion is straightforward.

High-accuracy second-order renormalized fluctuation expansion

Next, we point out the differences between the original second-order BM method of calculating D^r by Treloar and Masters, and our revised calculations within BM method. Approximations in the BM scheme are made in particular in two calculation steps. The first one is the truncation of the series in renormalized fluctuations to second order described in Eq. (S38). Secondly, also the matrices $d^{(0)}, d^{(2)}$ are approximated since they relate to infinite dimensional hydrodynamic matrices such as M, G, $G_{\langle \mathcal{M}_R \rangle}$, or $G^s_{\langle \mathcal{M}_R \rangle}$ which for a numerical evaluation must be truncated. In the works by BM on the hydrodynamic function and the high-frequency effective viscosity, and in the work by Treloar and Masters on rotational diffusion, due to technical difficulties severe truncations of these matrices have been made as discussed in detail in Ref.[8]. In our revised method, we also truncate the infinite dimensional hydrodynamic matrices, but different from earlier works an extrapolation to infinite dimension has been included. The details of this extrapolation procedure are along the lines described in Ref.[8] and are thus not repeated here.

S2 Hydrodynamic mobility matrices

We present here the details on how the hydrodynamic mobility matrices are calculated in the revised BM method.

The translational and rotational mobility matrices with tensor elements $\boldsymbol{\mu}_{ij}^{rr}(\mathbf{R}_1 \ldots \mathbf{R}_N)$ and $\boldsymbol{\mu}_{ij}^{tt}(\mathbf{R}_1 \ldots \mathbf{R}_N)$ appearing in Eqs. (5) and (S29) for the short-time rotational and translational self-diffusion coefficients, respectively, can be calculated using the hydrodynamic multipole matrices Z_0, μ_0, $\hat{Z}_0$, and G_0 introduced in Ref.[9,10]. Each of these matrices is indexed by the set of three indices (l,m,σ) with $l = 1,\ldots,\infty$, $m = -l, -l+1, \ldots, l$, and $\sigma = 0,1,2$. The matrix Z_0 is given by the formula

$$[Z_0(\mathbf{R}_i)]_{lm\sigma,l'm'\sigma'} = \delta_{ll'}\delta_{mm'}\eta(2a)^{2l+\sigma+\sigma'-1} z_{l,\sigma\sigma'} \,, \quad \text{(S39)}$$

where the dimensionless coefficients $z_{l,\sigma\sigma'}$ have been defined in Ref.[10]. The only non-vanishing elements of the matrix μ_0 are for $l = l' = 1$ and $\sigma = \sigma' = 0$, with

$$[\mu_0(\mathbf{R}_i)]_{lm0,l'm'0} = \delta_{ll'}\delta_{mm'}\frac{2}{9\eta a} \,, \quad \text{(S40)}$$

and for $l = l' = 1$ and $\sigma = \sigma' = 1$, with

$$[\mu_0(\mathbf{R}_i)]_{lm0,l'm'0} = \delta_{ll'}\delta_{mm'}\frac{1}{6\eta a^3} \,. \quad \text{(S41)}$$

The matrix $\hat{Z}_0$ is related to μ_0 and Z_0 by

$$\hat{Z}_0(\mathbf{R}_i) = Z_0(\mathbf{R}_i) - Z_0(\mathbf{R}_i)\mu_0(\mathbf{R}_i)Z_0(\mathbf{R}_i) \,. \quad \text{(S42)}$$

The Oseen tensor in multipole space, $G_0(\mathbf{R}_i,\mathbf{R}_j)$, is for non-overlapping configurations given by

$$[G_0(\mathbf{R}_i,\mathbf{R}_j)]_{lm\sigma,l'm'\sigma'} = \frac{n_{lm}}{\eta n_{l'm'}} S^{+-}(\mathbf{R}_i-\mathbf{R}_j,lm\sigma,l'm'\sigma'), \quad \text{for } |\mathbf{R}_i-\mathbf{R}_j|>2a, \tag{S43}$$

where the coefficients S^{+-} and n_{lm} have been introduced in Ref.[11]. The matrices noted above are used to construct the scattering series for the generalized multipole mobility matrix elements according to

$$\begin{aligned}
&\mu_{ij}(\mathbf{R}_1\dots\mathbf{R}_N)\\
&=\delta_{ij}\mu_0(\mathbf{R}_i)\\
&+(1-\delta_{ij})\,\mu_0(\mathbf{R}_i)Z_0(\mathbf{R}_i)G_0(\mathbf{R}_i,\mathbf{R}_j)Z_0(\mathbf{R}_j)\mu_0(\mathbf{R}_j)+\\
&+\sum_{\substack{k=1,\\ k\neq i,k\neq j}}^{N}\mu_0(\mathbf{R}_i)Z_0(\mathbf{R}_i)G_0(\mathbf{R}_i,\mathbf{R}_k)\hat{Z}_0(\mathbf{R}_k)\\
&\qquad\times G_0(\mathbf{R}_k,\mathbf{R}_j)Z_0(\mathbf{R}_j)\mu_0(\mathbf{R}_j)\\
&+\dots\,.
\end{aligned} \tag{S44}$$

In the multipole basis, the lowest multipole elements $l=1,\sigma=0$ correspond to translational motion. Therefore, the tensorial elements $\boldsymbol{\mu}^{tt}_{ij}$ of the translational mobility matrix can be calculated from the generalized mobility matrix with elements μ_{ij} by an appropriate projection. To this end, the projector $\overline{P}_t$ is introduced by

$$[\overline{P}_t]_{\alpha,lm\sigma}=\delta_{l1}\delta_{\sigma0}\sqrt{\frac{3}{4\pi}}[\mathbf{y}_{1m}]_\alpha\,, \tag{S45}$$

where $\alpha=1,2,3$ denote three Cartesian components. The tensor $\mathbf{y}_{1m}=X\mathbf{y}^{(R)}_{1m}$ is defined in Ref.[12], together with X and $\mathbf{y}^{(R)}$. Moreover, $\overline{P}_t^\dagger$ is the Hermitian conjugate of $\overline{P}_t$. Finally, the Cartesian mobility matrix is related to the generalized multipole mobility matrix by the projection operation

$$\boldsymbol{\mu}^{tt}_{ij}(\mathbf{R}_1\dots\mathbf{R}_N)=\overline{P}_t\mu_{ij}(\mathbf{R}_1\dots\mathbf{R}_N)\overline{P}_t^\dagger\,. \tag{S46}$$

The according expression for the rotational mobility matrix reads

$$\boldsymbol{\mu}^{rr}_{ij}(\mathbf{R}_1\dots\mathbf{R}_N)=\overline{P}_r\mu_{ij}(\mathbf{R}_1\dots\mathbf{R}_N)\overline{P}_r^\dagger\,. \tag{S47}$$

Here, the operator $\overline{P}_r$ projects from the multipole space the elements $l=1,\sigma=1$ to the Cartesian components corresponding to rotational motion, i.e.

$$[\overline{P}_r]_{\alpha,lm\sigma}=\delta_{l1}\delta_{\sigma1}\sqrt{\frac{3}{4\pi}}[\mathbf{y}_{1m}]_\alpha\,. \tag{S48}$$

In the scattering series for the mobility matrix in Eq. (S44), the single particle matrices μ_0Z_0 and $Z_0\mu_0$ at the start and end of a scattering sequence are different from the single-particle matrices $\hat{Z}_0$ dispersed in between the propagators G_0. In the derivation of the renormalized fluctuation expansion, it is thus very useful to rewrite the scattering series in a way that all the single particles matrices starting a scattering sequence or appearing in between the propagators G_0 are the same.

This can be achieved by introducing the matrix defined by[13]

$$M=\begin{bmatrix}\mu_0 & \mu_0Z_0\\ Z_0\mu_0 & \hat{Z}_0\end{bmatrix}. \tag{S49}$$

According to the above definition, the matrix M has in addition to the multipole indexes l,m,σ the index $u=1,2$. Therefore, $M_{1lm\sigma,1l'm'\sigma'}=[\mu_0]_{lm\sigma,l'm'\sigma'}$, $M_{1lm\sigma,2l'm'\sigma'}=[\mu_0Z_0]_{lm\sigma,l'm'\sigma'}$ et cetera. Similarly, we define the matrix $[G]_{ulm\sigma,u'l'm'\sigma'}$ by

$$G=\begin{bmatrix}0 & 0\\ 0 & G_0\end{bmatrix}, \tag{S50}$$

and generalize the projectors $\overline{P}_t$ and $\overline{P}_r$ to

$$[P_t]_{\alpha,ulm\sigma}=\delta_{u1}\delta_{l1}\delta_{\sigma0}\sqrt{\frac{3}{4\pi}}[\mathbf{y}_{1m}]_\alpha\,, \tag{S51}$$

$$[P_r]_{\alpha,ulm\sigma}=\delta_{u1}\delta_{l1}\delta_{\sigma1}\sqrt{\frac{3}{4\pi}}[\mathbf{y}_{1m}]_\alpha\,. \tag{S52}$$

In the extended space expressions in Eqs. (S44) and (S47), the rotational mobility matrix can be rewritten in the form given by Eq. (S1). This can be done analogously for the translational mobility matrix, with the only difference lying in the different projection operators.

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